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Interlayer correlations in ferromagnetic semiconductor superlattices EuS/PbS

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Abstract

Neutron diffraction and reflectivity data from all-semiconductor ferromagnetic/diamagnetic superlattices EuS/PbS provide direct evidence of antiferromagnetic coupling between EuTe layers across up to 90 Å of diamagnetic PbS, despite the absence of mobile carriers that can support long-range RKKY interactions. © 2001 Published by Elsevier Science B.V.

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Neutron diffraction and neutron reflectivity were employed for investigating the magnetism of EuS/PbS superlattices (SLs). EuS and PbS are closely lattice-matched ($a_{\mathrm{EuS}} = 5.97$ Å, $a_{\mathrm{PbS}} = 5.94$ Å) semiconducting compound of the NaCl type. PbS is nonmagnetic, and EuS is ferromagnetic (FM) with bulk $T_c = 16.6$ K. SL samples with [0 0 1] or [1 1 1] growth axis were prepared (on KCl or BaF_2, respectively) by thermal evaporation of PbS and electron-beam evaporation of EuS. The number of repeats was typically 20–30. The thickness of the EuS layers in different samples varied from 40 to 80 Å, and that of the PbS spacers from 10 to 100 Å.

Neutron scattering studies were performed at the NIST's Center for Neutron Research (NCRN), Gaithersburg, MD, USA. Diffraction measurements were carried out on a triple-axis spectrometer in elastic mode, with $\lambda = 2.35$ Å and angular collimation 40′ throughout. Regions in the vicinity of the principal Q-space points (corresponding to the average lattice periodicity in the SL structure) were thoroughly scanned in searching for magnetic scattering. Runs performed well above the bulk T_c showed only nuclear peaks at the expected positions accompanied by weak 'satellites' corresponding to the SL structure periodicity. Scans taken below T_c revealed new distinct components in the spectra. Examples of data obtained from transverse scan through the (0,2,0) reflection point in two different [0 0 1] specimens are displayed in Fig. 1. The origin of the additional low-T scattering components is undoubtedly magnetic. Their dependence on temperature closely follows the typical squared Brillouin function. Furthermore, the peaks – as discussed in greater detail below – show the expected response to external magnetic field B_{ext}. As can be seen in Fig. 1, there is no magnetic scattering at the reflection point: the two maxima are positioned half-way in between the central peak and the first-order nuclear satellites. Such a spectrum shape is a clear signature of *antiferromagnetic* (AFM) correlations between the magnetization in successive EuS layers.

The conclusion from the diffraction experiments are corroborated by the result of reflectometry studies. The Q-resolution in reflectivity measurements is much higher than in diffraction experiments. Reflectometry can only

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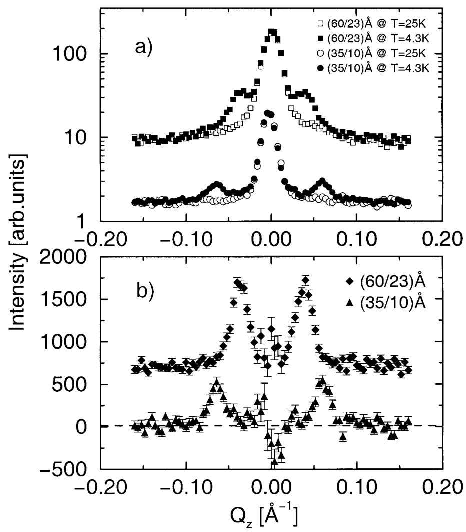

Fig. 1. Scans parallel to the SL growth axis (z) across the (0,2,0) reflecting points for (60/23) and (35/10) Å EuS/PbS specimens: (a) above and below T_c; (b) differential plots (purely magnetic scattering).

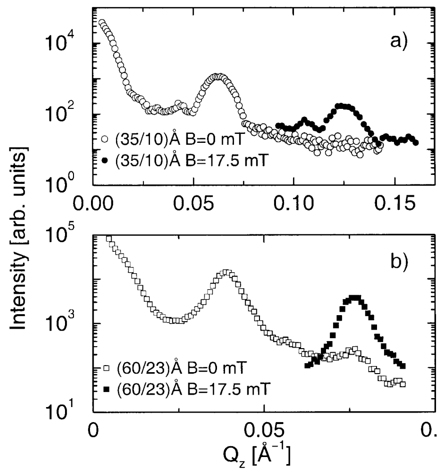

Fig. 2. Reflectivity profiles from the same two specimens used for obtaining the diffraction data in Fig. 1 at 4.3 K and zero and non-zero external magnetic field.

be used for investigating SL systems with *FM* layers. So far, all reported neutron scattering studies (see e.g. Refs. [1,2]) of magnetic semiconductor SLs have been done on systems prepared from AFM materials. Coupling across nonmagnetic spacers in such structures could not be probed by reflectivity. The present work on FM EuS/PbS is the first application of this powerful tool to an SL systems made exclusively of semiconducting materials.

Reflectivity measurements were done at NCNR on a vertical reflectometer using $\lambda = 4.75$ Å. Reflectivity profiles from two samples (the same ones used for obtaining the data in Fig. 1) are displayed in Fig. 2. Data taken at LHT show pronounced maxima at Q_z positions corresponding to doubled chemical SL period, a clear signature of AFM interlayer correlations. Such an AFM maximum was observed even for a system with the PbS spacer thickness D_{PbS} as large as 90 Å. After applying B_{ext} parallel to the layers, the AFM peak gradually decreases with increasing B_{ext} and completely disappears at 5–20 mT (depending on the D_{PbS} value). At the same time, a new peak emerges at the structural periodicity position, showing that B_{ext} causes a transition from the original AFM layer arrangement to a FM sequence. The AFM → FM transition was found to be almost completely reversible in the sample with $D_{PbS} = 10$ Å, but for thicker spacers the FM arrangement did not return to the AFM sequence after B_{ext} removal.

The above observations clearly indicate the existence of a significant AFM coupling between the FM EuS block across the non-magnetic spacers. Quantitative information about the coupling energy E_c, crucial for understanding the underlying physical mechanism, may be obtained from studying the AFM → FM transition. However, the data show that anisotropy energy (E_a) is another relevant factor in this process. The transition reversibility for smaller D_{PbS} values proves that in this region $E_c > E_a$; thus, the B_{ext} value needed to erase the AFM peak provides a measure of E_c. For larger D_{PbS}, on the other hand, E_a becomes dominant and the spins remain 'locked' in the FM configuration even after B_{ext} is removed. Here the AFM peak erasing field reflects only the E_a magnitude. Therefore, accurate determination of the $E_c(D_{PbS})$ curve from reflectivity data may require an elaborate analysis – and certainly will require the collection of more results from samples with different PbS spacer thicknesses.

In summary, neutron scattering experiments on EuS/PbS SLs reveal pronounced magnetic coupling between EuS blocks across $\geqslant 90$ Å of diamagnetic PbS. The exact physical mechanism of this phenomenon is yet to be understood. Theoretical models of interlayer exchange in metallic SL systems, which all rely on high concentrations of quasi-free electrons [3], cannot be applied to these all-semiconductor structures with low carrier concentrations (orders of magnitude lower than in metals!). The coupling sign oscillations, a 'trademark' of metallic SLs, are not observed in the EuS/PbS systems (AFM coupling was seen in the [0 0 1] specimens for all studied D_{PbS} values; in contrast, the [1 1 1] systems showed only FM coupling). The findings from the present work point to the need of a new theoretical insight. It should be stressed that interlayer coupling has also been observed in other all-semiconductor SL systems with AFM layers (e.g., MnTe/ZnTe [1], EuTe/PbTe [2]). It is worth noting that interlayer coupling in the AFM EuTe/PbTe multilayers has been the subject of a recent theoretical analysis based on a tight-binding approach [4]. The application of this formalism to the EuS/PbS system, whose electronic structure is closely related to that of EuTe/PbTe, would therefore be of great interest.

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